

REMARKS

By this response, claims 27 and 28 have been amended, and new claims 36-39 have been added, leaving claims 1-39 pending in the application. No new matter has been added by the amendments. It is respectfully submitted that the claim amendments (a) do not raise the issue of new matter, (b) do not raise any new issue that would require further search and/or consideration, and (c) place the application in better condition for appeal. Therefore, it is respectfully submitted that the amendments should be entered. Reconsideration and allowance are respectfully requested in view of the following remarks.

Allowable Subject Matter

Applicants gratefully acknowledge the indication in the Office Action that Claims 30 and 34 contain allowable subject matter. For reasons stated below, however, it is respectfully submitted that each of the pending claim is patentable.

Rejection Under 35 U.S.C. § 112, First Paragraph

Claim 27 stands rejected under 35 U.S.C. § 112, first paragraph, for the reasons stated at numbered paragraphs (1) and (2) on page 2 of the Office Action.

As acknowledged in the Office Action, the specification describes several examples falling within the claimed extinction of time range that have the mixture A. In light of this acknowledgment, Claim 27 has been amended to recite that the composition contains an effective amount of the mixture A. Applicants submit that the specification (a) provides a written description of the claimed subject matter and (b) the specification is enabling for the claimed subject matter.

More particularly, the specification describes three example compositions that fall within the scope of claim 27. In light of these working examples, Applicants submit that one skilled in the art would be able to make and use the claimed subject matter using the application disclosure as a guide. Applicants further submit that the Office Action has not provided adequate reasons to establish that one skilled in the art could not make and use other compositions that fall within the scope of claim 27 without undue experimentation. Thus, it is respectfully submitted that claim 27 complies with the requirements of 35 U.S.C. § 112, first paragraph. Therefore, withdrawal of the rejection is respectfully requested.

Rejection Under 35 U.S.C. § 112, Second Paragraph

Claims 28-35 stand rejected under 35 U.S.C. § 112, second paragraph, for the reasons stated at numbered paragraph (3) on page 2 of the Office Action.

Claim 28 has been amended to recite that “the composition contains an effective amount of d) and f) to enhance the arc-tracking and arc-erosion resistance properties of the article.” This amendment makes the body of claim 28 consistent with its preamble. As such, Applicants submit that this amendment makes explicit subject matter that was already implicit in claim 28 and thus does not raise any new issue. Thus, it is respectfully submitted that claims 28-35 comply with the requirements of 35 U.S.C. § 112, second paragraph. Therefore, withdrawal of the rejection is respectfully requested.

First Rejection Under 35 U.S.C. § 103

Claims 1-21, 26 and 27 stand rejected under 35 U.S.C. § 103(a) over U.S. Patent No. 5,543,450 to Takita et al. ("Takita") for the reasons stated at pages 3-4 of the Office Action. The rejection is respectfully traversed.

Takita discloses silicone rubber compositions that crosslink via a peroxide catalyst. See the paragraph bridging columns 4 to 5 of Takita. Such peroxide catalysts cannot be used together with the SiH group-containing component of the claimed composition. Hydrosilylation reactions involve the addition of Si-H bonds to double bonds, such as C=C with a Pt catalyst. Applicants submit that it is well known that the hydrosilylation reactions are sensitive to catalyst poisons. That is, elements such as N, P, S, Sn and As, even if present in trace amounts, can stop the reaction. As evidence of this fact, Applicants have attached a copy of each of U.S. Patent No. 5,416,147 (see column 1, lines 46-54 and column 1, last line to column 2, line 2); EP 0 604 104 A2 (see page 2, lines 19-22); U.S. Patent No. 6,303,728 (see column 1, lines 47-52) and Hoang Vi Tran, "Materials for Advanced Microlithography: Polymers for 157 nm Lithography and Acid Diffusion Measurements," page 51, second paragraph). As described in Tran, groups that are a good ligand for platinum poison the catalyst because they coordinate more strongly to the platinum center than the olefin.

In view of this well-known problem of catalyst poisoning, Applicants submit that it would not have been obvious to modify Takita's rubber composition that cures via a peroxide catalyst to result in a rubber composition that cures via a hydrosilylation reaction. As good ligands for platinum poison the catalyst, Applicants submit that one skilled in the art would not have been motivated to add to a rubber

composition that crosslinks via a hydrosilylation reaction any additive that interferes with this reaction, which is very sensitive to any trace chemical that interferes with platinum.

Also, the Office Action states that Takita teaches a mixture of cerium oxide and a platinum catalyst. Takita's compositions also contain a required azo compound in order to achieve the desired properties of the compositions. See, for example, column 2, lines 52-54, of Takita. Takita does not suggest that such nonflammability properties can be achieved without the addition of the azo compound. In contrast, the present specification describes examples of the claimed composition that have enhanced arc-tracking and arc-erosion resistance properties, but do not contain an azo compound, as required for Takita's compounds in order to have the needed nonflammability properties.

Accordingly, Applicants respectfully submit that claim 1 would not have been rendered obvious by Takita. Claims 2-15, 26 and 27, which depend from claim 1, also would not have been rendered obvious by Takita for at least the same reasons as those stated for claim 1. For example, the extinction of time value of less than 8 seconds recited in claim 27 is far below the values shown in Table 1 of Takita. Takita does not suggest any composition having the claimed extinction of time value, much less that Takita's composition could be optimized to achieve the claimed value.

Applicants further submit that the combinations of features recited in independent claims 16, 18 and 20 are also patentable over Takita for at least the same reasons as those discussed above with respect to claim 1. Claims 17 and 19 are also patentable for at least the same reasons as claims 16 and 18, respectively.

Therefore, withdrawal of the rejection is respectfully requested.

Second Rejection Under 35 U.S.C. § 103

Claims 1-25 and 27 stand rejected under 35 U.S.C. § 103(a) over JP 50-97644 ("JP '644") in view of U.S. Patent No. 4,110,300 to Matsushita ("Matsushita") for the reasons stated at numbered paragraph (6) on page 4 of the Office Action. The rejection is respectfully traversed.

JP '644 discloses a self-extinguishing silicone rubber composition. The composition comprises diorganopolysiloxane rubber, reinforcing filler silica, organic peroxide, platinum compound and iron oxide.

Matsushita discloses organopolysiloxane rubber, finely powdered silica, platinum and finely powdered γ -type iron sesquioxide.

The composition recited in claim 1 can be used to enhance the arc-tracking and arc-erosion resistance properties of an article. As shown in Table 1 at page 27 of the specification, control composition 1, which did not contain mixture A, B or C, exhibited a high arc-induced weight loss. However, as shown by the results given for Examples 1 and 2, adding a combination of FeO and Fe₂O₃ significantly reduced such weight loss. In addition, the compositions of Examples 1 and 2 exhibited improved flame resistance as compared to control composition 1.

In contrast, neither JP '644 nor Matsushita suggests that both flame resistance and arc-tracking and arc-erosion resistance properties of an article can be enhanced by a composition consisting essentially of an effective amount of mixture A, B or C, and the composition D, as claimed. Neither of these references provides any suggestion that improving the flame resistance of its particular disclosed compositions also results in an improvement in the arc-tracking and arc-erosion

resistance properties of those same compositions, much less a significant improvement in the latter properties. As such, Applicants submit that one skilled in the art would not have looked to either of these references in attempting to solve the problems identified and solved by the claimed composition, because these references provide no guidance regarding these problems.

Accordingly, it is respectfully submitted that claims 1-25 and 27 are also patentable over the combination of JP '644 and Matsushita. Therefore, withdrawal of the rejection is respectfully requested. Therefore, withdrawal of the rejection is respectfully requested.

Third Rejection Under 35 U.S.C. § 103

Claims 28, 29, 31 and 32 stand rejected under 35 U.S.C. § 103(a) over U.S. Patent No. 4,699,813 to Cavezzan ("Cavezzan") in view of JP '644 for the reasons stated at numbered paragraph (7) on pages 4-5 of the Office Action. The rejection is respectfully traversed.

The Examiner acknowledges that Cavezzan fails to disclose or suggest the addition of FeO and Fe₂O₃ to the composition disclosed in Example 6. As such, the Office acknowledges that Cavezzan does not disclose or suggest that "the composition contains an effective amount of d) and f) to enhance the arc-tracking and arc-erosion resistance properties of the article," where "d" is a platinum complex and "f" is a combination of FeO and Fe₂O₃.

Applicants respectfully submit that JP '644 does not suggest modifying Cavezzan's composition to include FeO and Fe₂O₃, as recited in claim 28. There is no disclosure or suggestion in Cavezzan that the organopolysiloxane composition

should impart arc-tracking and arc-erosion resistance properties to an article comprising the composition. The Examiner's position appears to be that it would have been obvious to add FeO and Fe₂O₃ to any known composition for the reason that JP '644 adds these to its silicone rubber composition. However, the Office Action has not established that it would have been desirable to improve the flame resistance of Cavezzan's organopolysiloxane composition. For example, the Office Action has identified no disclosure in Cavezzan that the organopolysiloxane composition is used in an application in which enhanced flame resistance is needed, much less the enhanced arc-tracking and arc-erosion resistance properties recited in claim 1. Accordingly, Applicants respectfully submit that the applied combination of references does not support the alleged *prima facie* obviousness. Thus, claim 28 is patentable over the applied references.

Claims 29, 31 and 32 depend from claim 28 and thus are also patentable. Therefore, withdrawal of the rejection is respectfully requested.

Fourth Rejection Under 35 U.S.C. § 103

Claims 33 and 35 stand rejected under 35 U.S.C. § 103(a) over U.S. Patent No. 5,674,966 to McDermott et al. ("McDermott") in view of JP '644 for the reasons stated at numbered paragraph (7) on pages 4-5 of the Office Action. The rejection is respectfully traversed.

Regarding claim 33, the Examiner acknowledges that McDermott fails to disclose or suggest the addition of FeO and Fe₂O₃ to the composition disclosed in Example 6. As such, the Office acknowledges that McDermott does not disclose or

suggest that “the composition contains an effective amount of d) and f),” where “d” is a platinum complex and “f” is a combination of FeO and Fe₂O₃.

Applicants respectfully submit that JP '644 does not suggest modifying McDermott's composition to include FeO and Fe₂O₃, as recited in claim 33. There is no disclosure or suggestion in McDermott that the molding resin should impart arc-tracking and arc-erosion resistance properties to an article comprising them. Applicants submit that the Office Action has not established that it would have been desirable to improve the flame resistance of McDermott's resin. The Office Action has identified no disclosure in McDermott that the resin composition is used in an application in which enhanced flame resistance is needed, much less the enhanced arc-tracking and arc-erosion resistance properties recited in claim 33. Accordingly, Applicants respectfully submit that the applied combination of references does not support the alleged *prima facie* obviousness. Thus, claim 33 is patentable over the applied references.

Claim 35 depends from claim 33 and thus is also patentable. Therefore, withdrawal of the rejection is respectfully requested. Therefore, withdrawal of the rejection is respectfully requested.

New Claims

New Claims 36, 37, 38 and 39 depend from claims 1, 16, 18 and 20, respectively, and each recites, *inter alia*, that “mixture B consists of at least one of: constituents B1 + B2 where constituent B1 has the meaning of constituent A1 and constituent B2 is cerium (IV) oxide and/or hydroxide” (emphasis added). For

reasons discussed above, Takita, for example, does not suggest the claimed composition. Applicants submit that claims 36-39 are also patentable.

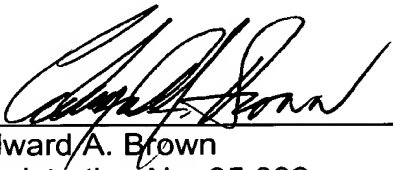
Conclusion

For the foregoing reasons, allowance of the application is respectfully requested. If there are any questions concerning this response, the Examiner is respectfully requested to contact the undersigned at the number given below.

Respectfully submitted,

BUCHANAN INGERSOLL PC (INCLUDING ATTORNEYS
FROM BURNS DOANE SWECKER & MATHIS)

Date: December 27, 2005

By: 
Edward A. Brown
Registration No. 35,033

P.O. Box 1404
Alexandria, Virginia 22313-1404
(703) 836-6620

D4

**Materials for Advanced Microlithography:
Polymers for 157 nm Lithography
and Acid Diffusion Measurements**

by

Hoang Vi Tran, B.S., M.A.

Dissertation

Presented to the Faculty of the Graduate School of
The University of Texas at Austin
in Partial Fulfillment
of the Requirements
for the Degree of

Doctor of Philosophy

**The University of Texas at Austin
May 2002**

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The proposed mechanism for platinum-catalyzed hydrosilylation, first brought forth by Chalk and Harrod in 1965, involves the sequence of steps outlined in Figure 3.5.^{74,78} Reduction of the catalyst by isopropanol converts the starting Pt^{IV} species to a Pt^{II} species. Coordination of the olefin provides the complex 3-1. Oxidative addition of HSiR_3 gives the hexacoordinated species 3-2, and migratory insertion of hydrogen onto the double bond changes the π complex to a σ complex 3-3. Reductive *syn* elimination results in the desired, hydrosilylated product.

During hydrosilylation, it is known that platinum is reduced: $\text{Pt}^{\text{IV}} \rightarrow \text{Pt}^{\text{II}} \rightarrow \text{Pt}^0 \rightarrow \text{Pt colloids}$. The formation of Pt colloids turns the reaction medium from a yellow color to a dark brown color, so one can visually observe the completion of the reaction. The platinum catalyst can tolerate a variety of functional groups, but groups that are good ligands for platinum (e.g., amines, phosphines, and sulfoxides) poison the catalyst because they coordinate more strongly to the platinum center than the olefin. It has also been observed that the presence of O_2 accelerates the reaction (O_2 is a co-catalyst).⁷⁹ As with radical hydrosilylation, terminal olefins react faster than internal olefins in platinum-catalyzed hydrosilylation (steric factor). Electron-rich olefins react more rapidly than electron-poor olefins (electronic factor). Hydrosilanes with electron-withdrawing groups react more rapidly than hydrosilanes with electron-donating

⁷⁸ Chalk, A. I.; Harrod, J. F. *J. Am. Chem. Soc.* 1965, 87, 16.

⁷⁹ Brook, M. A. *Formation of Si-C Bonds. The Synthesis of Functional Organosilanes*, In *Silicon in Organic, Organometallic, and Polymer Chemistry*, Wiley: New York, 2000; Chap. 12.



⑫

EUROPEAN PATENT APPLICATION

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㉖ Applicant : **SHIN-ETSU CHEMICAL CO., LTD.**
6-1, Ohtemachi 2-chome
Chiyoda-ku Tokyo (JP)

㉗ Inventor : **Yoshikawa, Yuji**
1610 Nishikamiisobe
Annaka-shi, Gunma-Ken (JP)
Inventor : **Yamamoto, Kenji**
12-37 Isobe 3-chome
Annaka-shi, Gunma-ken (JP)
Inventor : **Tanaka, Kouichi**
2-32, Isobe 4-chome
Annaka-shi, Gunma-ken (JP)
Inventor : **Takarada, Mitsuhiro**
279-6 Kenzaki-machi
Takasaki-shi, Gunma-ken (JP)

㉘ Representative : **Stoner, Gerard Patrick et al**
MEWBURN ELLIS
York House
23 Kingsway
London WC2B 6HP (GB)

㉙ **Hydrosilylative compositions and hydrosilylation reaction processes.**

㉚ A blend of (A) a silicone compound having at least two alkenyl or alkynyl groups in a molecule, (B) an organic silicon compound having at least two hydrogen atoms each directly attached to a silicon atom in a molecule, and (C) a hydrosilylation catalyst is blended with (D) an organic iron or aluminum compound to form a hydrosilylative composition. Component (D) scavenges a catalyst poison, permitting components (A) and (B) to undergo hydrosilylation reaction with the aid of catalyst (C).

This invention relates to a hydrosilylative composition comprising a silicone compound having an aliphatic unsaturated bond, a compound having a silyldyne radical and a hydrosilylation catalyst such as platinum and a hydrosilylation reaction process of subjecting the former two compounds to hydrosilylation reaction in the presence of the catalyst. More particularly, it relates to a hydrosilylative composition and process which permit hydrosilylation reaction to readily proceed even in a system which contains or contacts with catalyst poisons.

It is well known that silicone compounds having an aliphatic unsaturated bond and compounds having a silyldyne (-SiH) radical undergo hydrosilylation reaction in the presence of hydrosilylation catalysts such as platinum or rhodium series catalysts whereby curing takes place through addition of the silyldyne radical to the unsaturated bond. There were proposed in the art a variety of curable resin compositions which cure through such hydrosilylation reaction.

Since the hydrosilylation reaction is high in reactivity, curing of resin compositions through hydrosilylation reaction has the advantage that the compositions cure uniformly both at the surface and the interior and with a minimal volume loss. Another advantage is that an overall process from mixing of components to form a composition to the end of curing is energy saving.

Owing to these advantages, coupled with availability in liquid form and ease of handling without a need for special equipment or techniques, the curable resin compositions now find widespread use in a variety of fields. For example, these compositions are used for electrical insulation, mold patterning, junction coating, optical fibers, LIMS molding, silicone gel, release paper, and tackifiers.

However, the hydrosilylation reaction is sensitive to catalyst poisons. More particularly, if such elements as N, P, S, Sn and As, even in trace amounts, are present in the compositions, or if such elements are present in substrates to which the compositions are coated and cured, then these elements become catalyst poisons to considerably inhibit hydrosilylation reaction. Retarded hydrosilylation reaction can result in short curing.

It is then a common practice to carry out hydrosilylation reaction while utilizing suitable catalyst poison scavenging means. That is, catalyst poisons can be removed from the composition itself or the substrate to which the composition is coated and cured, for example, by pretreating the components of the composition with activated carbon or by heat cleaning the substrate to pyrolytically decomposing off the catalyst poisons. It is also possible to carry out hydrosilylation reaction by raising the reaction temperature to enhance the activity of the hydrosilylation catalyst or by increasing the amount of the hydrosilylation catalyst added.

These approaches, however, are disadvantageous both in process and economy. It is thus desired to ensure that hydrosilylation reaction proceeds even in the presence of trace amounts of catalyst poisons.

A general aim herein is to provide novel hydrosilylation compositions and methods of using and preparing them, preferably compositions and processes which permit hydrosilylation reaction to proceed without inhibition even in the presence of trace amounts of catalyst poisons including such elements as N, P, S, Sn and As.

Regarding the hydrosilylation reaction between a silicone compound having at least two alkenyl or alkynyl groups in a molecule and an organic silicon compound having at least two hydrogen atoms each directly attached to a silicon atom in a molecule in the presence of a hydrosilylation catalyst, the inventors have found that the poisoning of a catalyst poison component containing N, P, S, Sn or As is inhibited if organic iron compounds such as iron octylate, iron naphthenate and iron acetylacetonate or organic aluminum compounds, for example, aluminum alkoxides such as aluminum butoxide, aluminum isobutoxide, and aluminum isopropoxide and aluminum chelates such as acetylacetonatoaluminum and ethylacetoacetonatoaluminum are present in the reaction system. Then even when such a catalyst poison component is contained in a composition or a substrate with which the composition comes in contact, hydrosilylation reaction can take place relatively freely. Thus, broadly the invention provides the use of such iron and/or aluminium compounds to inhibit catalyst poisoning in this context.

One specific aspect provides a hydrosilylative composition comprising

(A) a silicone compound having at least two alkenyl or alkynyl groups in a molecule,

(B) an organic silicon compound having at least two hydrogen atoms each directly attached to a silicon atom in a molecule,

(C) a hydrosilylation catalyst, and

(D) at least one of an organic iron compound and an organic aluminum compound.

In a second aspect, the present invention provides a hydrosilylation reaction process comprising the step of:

subjecting (A) a silicone compound having at least two alkenyl or alkynyl groups in a molecule and (B) an organic silicon compound having at least two hydrogen atoms each directly attached to a silicon atom in a molecule to hydrosilylation reaction in the presence of (C) a hydrosilylation catalyst, while using (D) at least one of an organic iron compound and an organic aluminum compound in the reaction system.

It is not well understood why the addition of an organic iron or aluminum compound permits hydrosilylation

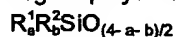
reaction to take place even in the presence of a catalyst poison component containing N, P, S, Sn or As. Though not bound by theory, we hypothesise as follows. If a catalyst poison is present, a Group VIII transition metal catalyst as the hydrosilylation catalyst coordinates with the catalyst poison so that hydrosilylation reaction proceeds no longer. Nevertheless, if hydrosilylation reaction is effected in a system where a Group VIII transition metal catalyst coexists with an organic iron or aluminum compound, the organic iron or aluminum compound preferentially coordinates with the catalyst poison so that the Group VIII transition metal catalyst may remain active. Then even in the presence of a catalyst poison, hydrosilylation reaction can proceed in the same manner as does a catalyst poison - free system.

The invention also embraces precursor blend compositions such as (A), (B) and (D); (A), (C) and (D), and (B), (C) and (D).

The hydrosilylative composition according to the first aspect of the present invention can be prepared as a curable resin composition and includes (A) a silicone compound having at least two alkenyl or alkynyl groups in a molecule, (B) an organic silicon compound having at least two hydrogen atoms each directly attached to a silicon atom in a molecule, and (C) a hydrosilylation catalyst.

Component (A) is a silicone compound or organopolysiloxane which is not particularly limited insofar as it has at least two alkenyl or alkynyl groups in a molecule. Exemplary alkenyl group include aliphatic unsaturated hydrocarbon groups such as vinyl, allyl, methylvinyl, propenyl, butenyl, pentenyl and hexenyl groups, and cyclic unsaturated hydrocarbon groups such as cyclopropenyl, cyclobutenyl, cyclopentenyl and cyclohexenyl groups. Exemplary alkynyl groups include ethynyl, propargyl and α , α -dimethylpropargyl groups.

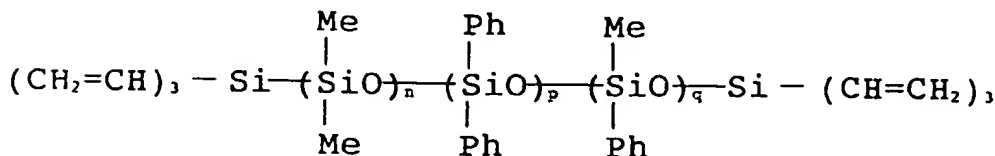
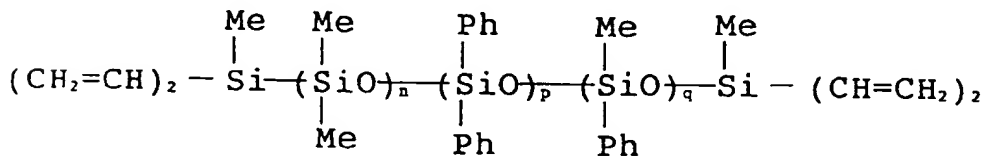
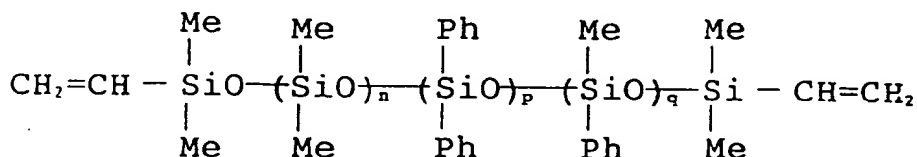
More detailedly, the silicone compound or organopolysiloxane may typically have the following formula:



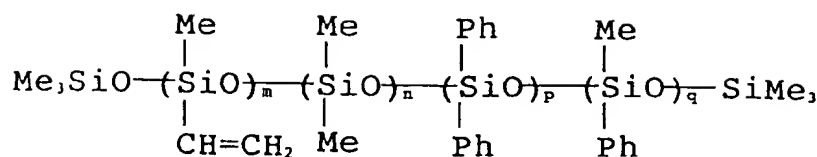
wherein R¹ represents an alkenyl or alkynyl group, R² represents a monovalent hydrocarbon group having 1 to 10 carbon atoms (but not alkenyl or alkynyl group) and the letters a and b represent positive numbers satisfying the ranges of $0 < a < 4$, $0 < b < 4$ and $0 < a + b < 4$ so that the organopolysiloxane has at least two alkenyl or alkynyl groups.

The monovalent hydrocarbon groups of R² includes an alkyl groups such as methyl, ethyl, propyl, butyl and the like, an aryl group such as phenyl and the like, an aralkyl group such as benzyl and the like, an cycloalkyl group such as cyclohexyl and the like. They may have substitution, provided that the substitution does not spoil the curability of the composition or any other conventionally-desired property. For example, alkyl may be halo-substituted alkyl such as trifluoropropylalkyl, or the like. Unsubstituted methyl and phenyl are preferred.

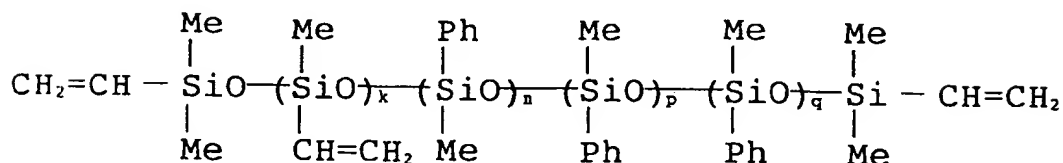
Examples of suitable polysiloxanes are as follows.



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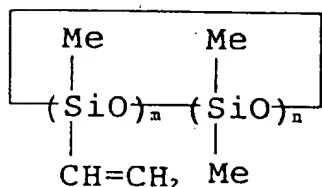


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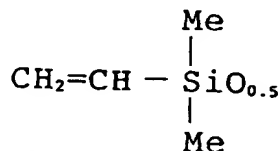


wherein Me is methyl, Ph is phenyl, $n \geq 0$, $p \geq 0$, $q \geq 0$, $m \geq 2$ and $k \geq 0$.

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Also included are vinyl group-containing MQ resins which are copolymers of

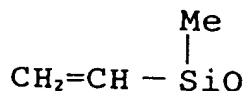
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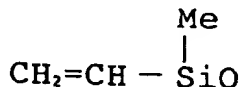
units and SiO_2 units, silicone resins containing $\text{CH}_2=\text{CHSiO}_{1.5}$ units, silicone resins containing $\text{CH}_2=\text{CHSiO}_{1.5}$ units and Me_2SiO units, silicone resins containing

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units and $\text{MeSiO}_{1.5}$ units, and silicone resins containing

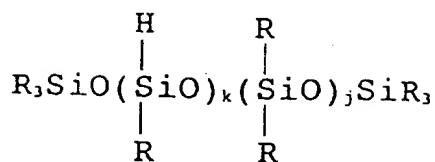
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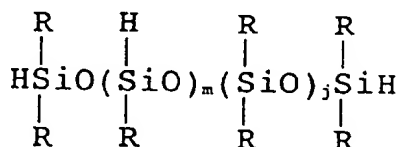
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units and $\text{C}_6\text{H}_5\text{SiO}_{1.5}$ units.

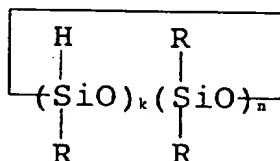
Component (B) is an organic silicon compound which is not particularly limited insofar as it has at least two hydrogen atoms each directly attached to a silicon atom in a molecule. Exemplary are organohydrogensilanes and organohydrogensiloxanes. The organohydrogensiloxanes may be linear, cyclic or network and include from oligomers to polymers although tetrasiloxane and higher ones having a viscosity of up to 10,000 centipoise at room temperature are especially preferred when the flexibility of cured coatings and workability are taken into account. Typical structural formulae for component (B) are as follows.



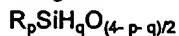
R is a monovalent hydrocarbon group, preferably an alkyl, cycloalkyl, aralkyl or aryl group having 1 to 8 carbon atoms. R may be unsubstituted, or substituted provided that the substituent does not spoil the curability of the compositions or other conventionally-desired property. Unsubstituted methyl or phenyl groups are preferred. k and j are integers; $k \geq 2$ and $j \geq 0$.



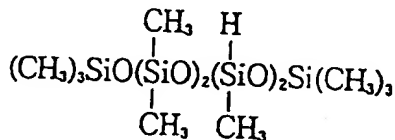
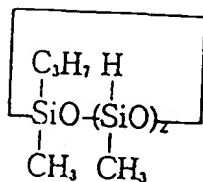
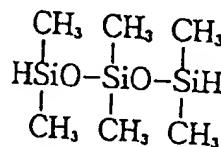
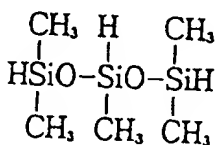
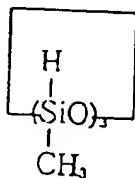
R and j are as defined above, and m is an integer of $m \geq 0$.

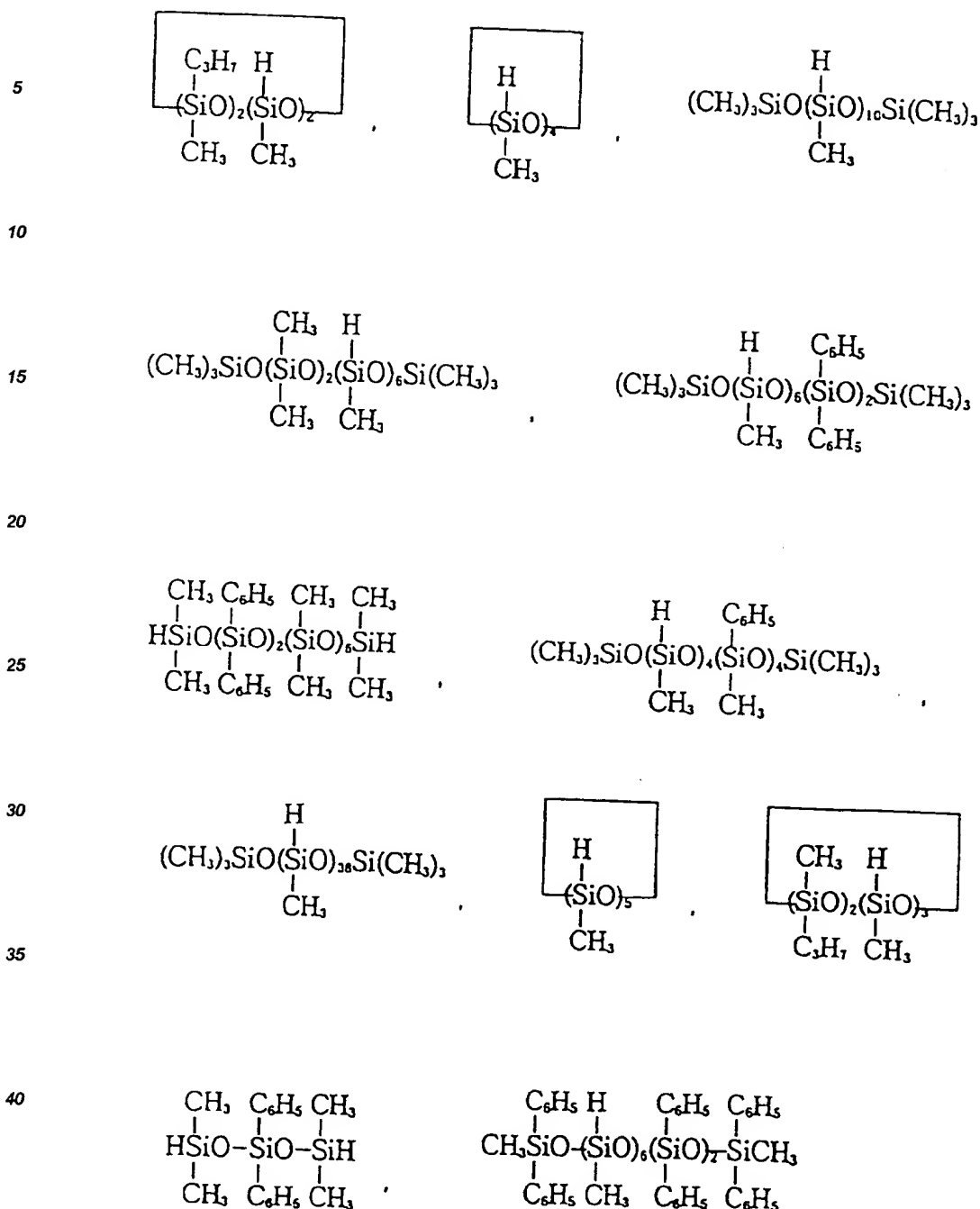


R and k are as defined above, and n is an integer of $n \geq 1$.



R is as defined above, and p and q are numbers to satisfy $0 < p + q < 2$, $0 < p < 2$, and $0 < q < 2$. Illustrative, non-limiting examples of component (B) are shown below.





Components (A) and (B) are preferably blended such that the molar ratio of alkenyl/alkynyl group in component (A) to the hydrogen atom in component (B) may range from 1:0.5 to 1:50, especially from 1:1 to 1:10. On this basis, if the proportion of component (B) is less than 0.5, crosslinking may be short. If the proportion of component (B) is more than 50, unreacted component (B) may bleed on a coating.

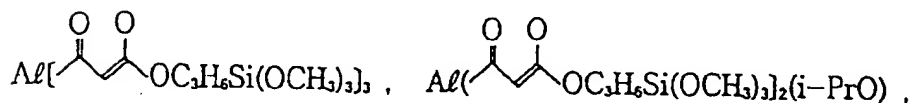
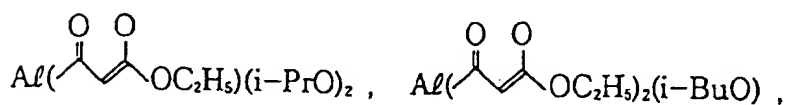
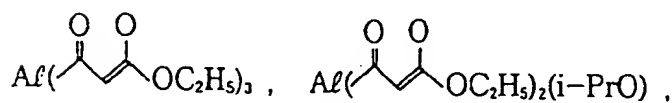
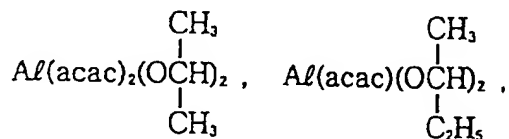
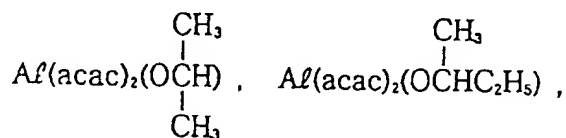
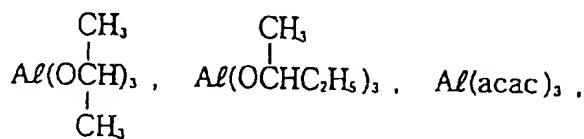
Component (C) is a hydrosilylation catalyst which is generally selected from transition metals of Group VIII in the Periodic Table and their compounds. Exemplary Group VIII transition metal compounds are complexes of platinum, palladium, rhodium and ruthenium, for example, such as PtCl_4 , $\text{H}_2\text{PtCl}_6 \cdot 6\text{H}_2\text{O}$, Pt-ether complexes, Pt - olefin complexes, $\text{PdCl}_2(\text{PPh}_3)_2$, $\text{PdCl}_2(\text{PhCN})_2$, $\text{RhCl}_2(\text{PPh}_3)_3$ wherein Ph is phenyl. Other known hydrosilylation catalysts and mixtures of such catalysts are also useful. If desired, these catalysts may be used by diluting with solvents such as alcohol, aromatic, hydrocarbon, ketone and basic solvents.

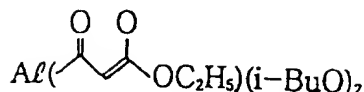
Component (C) is blended in an amount to give 0.1 to 10,000 parts by weight of metal per million parts by weight of component (B). Catalysts may be insufficient with less than 0.1 ppm of component (C) whereas more than 10,000 ppm of component (C) may cause the cured coating to be colored due to the catalyst's own

color and is economically disadvantageous since the Group VIII metals are expensive.

Catalyst poison controlling component (D) in the form of an organic iron or aluminium compound is used with, e.g. added to a blend of, components (A), (B) and (C).

The organic iron and aluminum compounds as component (D) are not particularly limited insofar as they are soluble in components (A) and (B). Exemplary organic iron compounds include ferric organosalts e.g. ferric octylate, ferric acetate, ferric propionate and ferric naphthenate, and acetyl-acetonatoiron. Exemplary organic aluminium compounds are aluminum alkoxides and chelates as shown below.

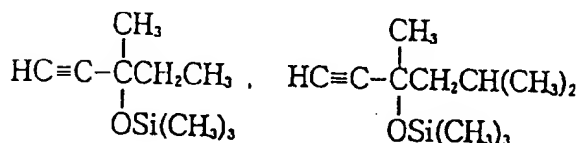
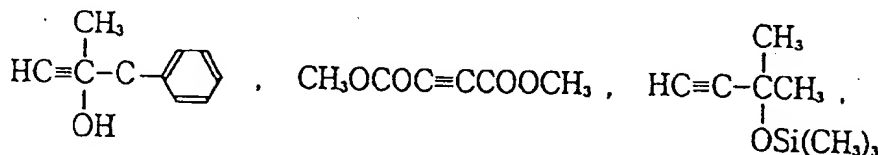
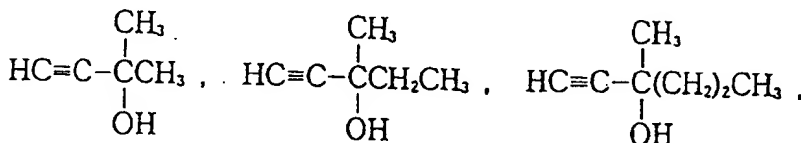




In the formulae, acac is acetylacetonato, Pr is propyl, and Bu is butyl.

Component (D) may be included in an amount corresponding to the content of catalyst poison envisaged. Since even trace amounts of catalyst poison can affect hydrosilylation reaction or catalyst component (C), component (D) is preferably used in a weight of 10 to 10,000 times the weight of the metal of component (C) in order to prohibit any such influence. Less than 10 times the metal weight of component (D) may provide insufficient guard against the catalyst poison whereas more than 10,000 times the metal weight of component (D) may cause the cured coating to be cracked or colored with iron.

In addition to the essential components (A) to (D), the composition of the present invention may further include an agent for controlling hydrosilylation reaction for the purpose of adjusting ease of operation. Such control agents include acetylene alcohols, siloxane-modified acetylene alcohols, methylvinylcyclotetrasiloxane, and hydroperoxides. Their illustrative examples are given below. Such agents are known.



These control agents may be added in any desired amount in accordance with desired operating conditions, typically in an amount of 1 to 1,000% of the metal weight of component (C).

Also if desired, reinforcing fillers may be used in the compositions.

Exemplary fillers are reinforcing silica, ground quartz, iron oxide, alumina and vinyl-containing silicone resins.

Various other additives may be used in accordance with the intended use of the composition. Examples include heat resistance modifiers such as oxides and hydroxides of cerium and iron; coloring agents such as organic pigments or dyes and inorganic pigments (e.g., TiO_2 , Fe_2O_3 , and C); tackifiers such as carbon functional silanes and siloxanes; mold release agents such as silicone fluid, raw rubber and metal fatty acid salts; flame retardants such as zinc carbonate and calcium carbonate; flow control agents; and anti-settling agents.

Depending on operating conditions, the composition of the invention may be used without solvent. Where component (A) or (B) is highly viscous or component (A) is solid, it may be dissolved in an organic solvent. The organic solvent used herein is not particularly limited insofar as components (A) to (D) are soluble therein. Exemplary solvents are aromatic hydrocarbons such as benzene, toluene and xylene; aliphatic hydrocarbons such as hexane, heptane and octane; ketones such as acetone, methyl ethyl ketone, cyclohexanone, and methyl isobutyl ketone; esters such as ethyl acetate, butyl acetate and isobutyl acetate; and haloalkanes such as 1,1,1-trichloroethane, trichloroethylene and methylene chloride. Toluene and xylene are preferred for industrial purposes.

The procedure of preparing the composition of the invention is not limitative. For example, components (A), (C) and (D) are first dissolved in an organic solvent and the solution is then mixed with component (B).

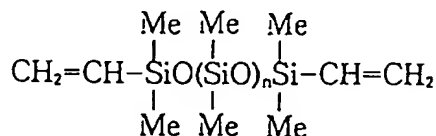
Hydrosilylation reaction will take place in the composition of the invention under conventional conditions, typically by heating at a temperature of 50 to 150°C.

EXAMPLE

Examples of the present invention are given below by way of illustration and not by way of limitation. All parts are by weight.

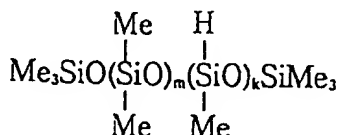
Example 1

The following four components were mixed. Note that Me is methyl.
(1) vinyl-containing silicone compound, 60 parts



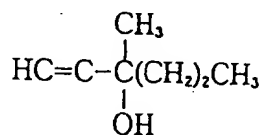
viscosity = 10,000 centistokes

(2) hydrogen atom-containing organic silicon compound, 10 parts



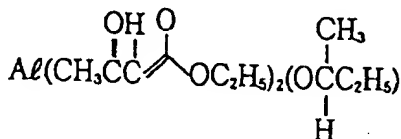
(M ≅ 12, k ≅ 12)

(3) reaction control agent, 0.3 parts



(4) xylene, 30 parts

The mixture was further blended with 2 parts of a xylene solution of chloroplatinic acid (Pt content 0.05% by weight), 2 parts of a 50% toluene solution of an aluminum chelate of the formula:



(Kelope ACS commercially available from Hope Pharmaceutical K.K.), and 100 parts of xylene to form a solution A. Solution A, 100 parts, was thoroughly mixed with 0.025 parts of N-(β-aminoethyl)-γ-aminopropyltrimethoxysilane as a nitrogenous compound. The resulting composition was cast over an iron plate, air dried for 10 minutes, and baked at 150°C for 5 minutes to form a cured coating. This cured coating had physical properties as shown in Table 1.

Example 2

Solution A obtained in Example 1 was coated onto a cured acryl-melamine coated iron plate by means of

a bar coater, air dried for 10 minutes, and baked at 150°C for 5 minutes to form a cured coating. This cured coating had physical properties as shown in Table 1.

Example 3

A solution B was prepared by the same procedure as solution A in Example 1 except that 2 parts of a 50% toluene solution of ferric octylate (Octope Fe commercially available from Hope Pharmaceutical K.K.) was added instead of 2 parts of the aluminum chelate toluene solution. Then following the procedure of Example 1, a cured coating was formed from solution B. This cured coating had physical properties as shown in Table 1.

Comparative Example 1

A solution C was prepared by mixing 100 parts of the addition type silicone varnish used in Example 1, 2 parts of the platinum catalyst solution used in Example 1, and 100 parts of xylene. Solution C, 100 parts, was thoroughly mixed with 0.025 parts of N-(β -aminoethyl)- γ -aminopropyltrimethoxysilane. The resulting composition was cast over an iron plate, air dried for 10 minutes, and baked at 150°C for 5 minutes to form a cured coating. This cured coating had physical properties as shown in Table 1.

Comparative Example 2

Solution C obtained in Comparative Example 1 was coated onto a cured acryl-melamine coated iron plate by means of a bar coater, air dried for 10 minutes, and baked at 150°C for 5 minutes to form a cured coating. This cured coating had physical properties as shown in Table 1.

Comparative Example 3

As in Comparative Example 1, the coating was baked at 180°C for 15 minutes. This cured coating had physical properties as shown in Table 1.

Comparative Example 4

A cured coating was formed by the same procedure as in Comparative Example 1 except that 10 parts of the platinum catalyst solution was added to solution C. This cured coating had physical properties as shown in Table 1.

Table 1

	Dryness	Solvent resistance
Example 1	tack-free	OK
Example 2	tack-free	OK
Example 3	tack-free	OK
Comparative Example 1	tacky	dissolved
Comparative Example 2	tacky	dissolved
Comparative Example 3	tack-free	whitened
Comparative Example 4	tack-free	OK

Dryness was examined by touching the coating with fingers.

Solvent resistance was examined by rubbing the coating with xylene-impregnated cotton wadding 10 strokes under a load of about 500 grams and then visually observing the outer appearance.

Component (D) permits components (A) and (B) to undergo hydrosilylation reaction with the aid of a hydrosilylation catalyst even in the presence of a catalyst poison.

Although some preferred embodiments have been described, many modifications and variations may be made thereto in the light of the above teachings. It is therefore to be understood that within the scope of the appended claims, the invention may be practiced otherwise than as specifically described.

Claims

1. A hydrosilylative composition comprising
- 5 (A) a silicone compound having at least two alkenyl or alkynyl groups in a molecule,
(B) an organic silicon compound having at least two hydrogen atoms each directly attached to a silicon atom in a molecule,
(C) a hydrosilylation catalyst, and
(D) at least one of an organic iron compound and an organic aluminum compound.
- 10 2. A hydrosilylation reaction process comprising the steps of:
subjecting (A) a silicone compound having at least two alkenyl or alkynyl groups in a molecule and
(B) an organic silicon compound having at least two hydrogen atoms each directly attached to a silicon atom in a molecule to hydrosilylation reaction in the presence of (C) a hydrosilylation catalyst, and
15 adding (D) at least one of an organic iron compound and an organic aluminum compound to the reaction system.
3. The process of claim 2 wherein the hydrosilylation reaction is effected in a system containing an element selected from the group consisting of N, P, S, Sn and As or in a system in contact with a material containing said element.
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